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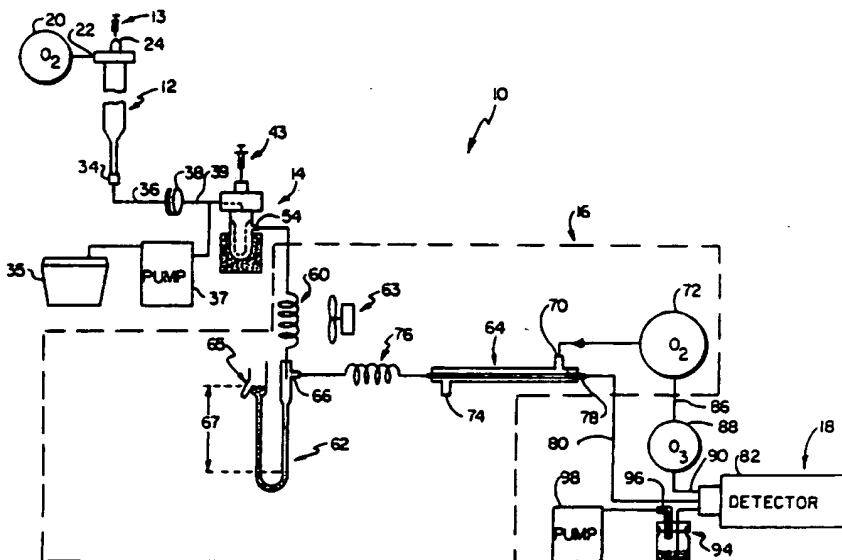
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(54) Title: APPARATUS AND METHOD FOR MEASURING NITROGEN CONTENT IN AQUEOUS SYSTEMS

(57) Abstract

A method and apparatus for determining Total Nitrogen Content and/or Combined Nitrate-Nitrite Nitrogen Content in an aqueous system is disclosed. To measure the Total Nitrogen Content, an oxidation tube (12) having an oxidation catalyst (32) receives a carrier gas (20) including oxygen and a sample (13) of the aqueous system. The oxidation tube (12) converts the aqueous sample (13) to a product gas including nitrogen dioxide. The nitrogen dioxide is then converted to nitric oxide in a reaction chamber (14) having a liquid chemical reduction agent (41). The nitric oxide is received and measured to determine the Total Nitrogen Content by a detection apparatus (18). To measure the Combined Nitrate-Nitrite Nitrogen Content in the aqueous system, a second aqueous sample (43) is injected into the reaction chamber (14) and converted to a second quantity of nitric oxide. The second quantity of nitric oxide is received and measured by the detection apparatus (18).



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APPARATUS AND METHOD FOR MEASURING NITROGEN CONTENT IN AQUEOUS SYSTEMS

BACKGROUND OF THE INVENTION

The present invention relates to a system for measuring nitrogen
5 content. More particularly, the present invention provides an analytical method
and apparatus for measuring nitrogen content in aqueous samples such as waste
water.

As used herein, the following definitions are
intended.

10 Total Combined Nitrogen Content means the
amount of nitrogen existing with other chemical
elements in certain organic and
inorganic compounds.

15 Total Kjeldahl Nitrogen means the amount of
nitrogen existing in certain organic compounds and
in ammonia and ammonium compounds.

20 Nitrite Nitrogen means nitrogen existing in
inorganic compounds containing the nitrite (NO_2^-)
ion.

25 Nitrate Nitrogen means nitrogen existing in
inorganic compounds containing the nitrate (NO_3^-)
ion.

30 Combined Nitrate-Nitrite Nitrogen Content is the
amount of nitrogen existing in a mixture of nitrites
and nitrates without distinguishing from the parent
species.

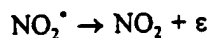
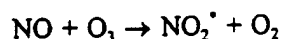
NO_x means gaseous oxides of nitrogen.

35 Quantitatively determining the amount of nitrogen in a mixture is
among the most important of analytical measurements. In waste water treatment

processes, nitrogen enters the water in human and animal excrement as organic compounds such as, for example, via proteins, peptides and amino acids. Nitrogen can also enter the water from organic and inorganic forms from industrial wastes. In biological treatment plant processes, the organic nitrogen
5 is converted by bacterial action to ammonia nitrogen, while other bacteria convert the ammonia nitrogen to nitrite nitrogen and finally then to nitrate nitrogen. Nitrates are not desirable in effluent waters because nitrates are nutrients for algal growth.

A commonly known procedure for analysis of organically-bound
10 nitrogen is the Kjeldahl method. In the Kjeldahl method, the sample is heated in concentrated sulfuric acid containing a catalyst. This procedure converts the nitrogen to ammonia. On the addition of sodium hydroxide followed by boiling, ammonia is distilled into a dilute boric acid solution, which is then titrated with acid.

15 More recently, chemiluminescent detection equipment for detecting organically-bound nitrogen has been developed. These chemiluminescent detectors are based on the reaction of nitric oxide with ozone to form metastable or excited nitrogen dioxide (NO_2^*). Almost instantaneously, the excited nitrogen dioxide relaxes to its ground state with a resulting photo-emission (ϵ), as
20 represented by the following reactions:



The chemiluminescent nitrogen detectors are commonly known as
25 NO/ NO_x detectors and are used to detect ambient NO and NO_x concentrations in the air. Such NO/ NO_x concentration detectors are presently made by several companies, but they are limited in that they are able to detect nitrogen only as NO/ NO_x gases in gaseous samples. Alone, they cannot be utilized with liquid or solid organic compound samples.

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A chemiluminescent nitrogen detection apparatus and method for samples in a phase other than gas has been disclosed in U.S. Patent 4,018,562 issued to Parks et al. Parks et al. discloses a system including an inert gas, such as helium or argon, a source of oxygen, a furnace or other pyrolyzing means
5 connected to receive the carrier gas (helium or argon), the oxygen and a sample containing nitrogen. In order to determine the Total Combined Nitrogen Content, the furnace pyrolyses the sample to convert the sample to oxides of nitrogen. The product gases from the furnace are then provided to a dryer. A reaction chamber of a chemiluminescent detector is connected to an output of the
10 dryer. An ozone generator is connected to the reaction chamber and generates ozone from oxygen received from the oxygen source. The reaction chamber receives the gases from the dryer and the ozone from the ozone generator. The ozone and nitric oxide mix within the reaction chamber and react, as described above, resulting in the photo-emission of light. The chemiluminescent detector
15 receives the emitted light and provides an electrical signal proportional to the nitric oxide, which is also proportional to the Total Combined Nitrogen Content.

SUMMARY OF THE INVENTION

The present invention provides an apparatus and method for determining Total Combined Nitrogen Content and/or Combined Nitrate-Nitrite
20 Nitrogen Content in a liquid system. The apparatus comprises an oxidation tube, a nitrate-nitrite reaction chamber, a dryer and a detection apparatus. To measure the Total Combined Nitrogen Content, the oxidation tube having an oxidation catalyst receives a carrier gas including oxygen and a sample from the liquid system. The oxidation tube converts the liquid sample to a product gas that
25 includes nitrogen dioxide and nitric oxide. The nitrogen dioxide is then converted to nitric oxide in the reaction chamber. The reaction chamber includes a liquid chemical reduction agent, preferably vanadium and sulfuric acid. The nitric

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oxide is provided to the dryer where the nitric oxide is dried sufficiently for the detection apparatus, such as a chemiluminescent detector.

In addition, the present system can be used to determine the Combined Nitrate-Nitrite Nitrogen Content in the liquid system. When operated to determine the Combined Nitrate-Nitrite Nitrogen Content, a second liquid sample is injected into the reaction chamber and converted to a second portion of nitric oxide. The nitric oxide is again provided to the dryer for drying. The second portion of nitric oxide is received and then measured by the detection apparatus to determine the Combined Nitrate-Nitrite Nitrogen Content.

10 BRIEF DESCRIPTION OF THE DRAWINGS

Figure 1 is a mechanical schematic drawing of a nitrogen analyzer according to the present invention;

Figure 2 is a partial sectional view of an oxidation tube;

Figure 3 is a partial sectional view of a nitrate-nitrite reaction chamber; and

Figures 4A and 4B are side elevational views of the reaction chamber in operation.

DETAILED DESCRIPTION OF THE PREFERRED EMBODIMENTS

A nitrogen measuring system is indicated generally at 10 in Figure 1. The system 10 includes a catalytic oxidation (combustion) tube 12, a nitrate-nitrite ("N-N") reaction chamber 14, drying means indicated generally at 16, and a detector 18, such as a conventional gas chromatograph or a NO/NO_x chemiluminescent detection apparatus herein depicted. The system 10 quantitatively determines the Total Combined Nitrogen Content and the Combined Nitrate Nitrite Nitrogen Content in an aqueous system from different samples. From these determinations, the Total Kjeldahl Nitrogen can be calculated as the difference between Total Combined Nitrogen Content and Combined Nitrate-Nitrite Nitrogen Content.

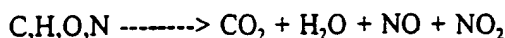
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The catalytic oxidation tube 12 is illustrated in detail in Figure 2. The catalytic oxidation tube has a first inlet 22 connected to an oxygen source 20 to supply metered oxygen. As will be described, oxygen is used throughout the measuring system 10 as the carrier gas. Therefore, it should be understood that applicable components of the system 10 are made from material suitable for carrying oxygen as well as NO_x to be described below.

To determine the Total Combined Nitrogen Content, an aqueous sample containing nitrogen is injected into the catalytic oxidation tube 12 with a microsyringe 13 through a second inlet 24 to pyrolyze the sample in an oxygen atmosphere to form oxides of nitrogen, mostly nitric oxide. For example, approximately 5-10% by volume of the product gas is typically nitrogen dioxide. The catalytic oxidation tube 12 is made from quartz and has an upper expansion zone 26 to minimize the pressure increase due to evaporation of water and to keep the evaporated sample in the oxidation tube 12. At a lower end 28 of the oxidation tube 12, the tube 12 is narrowed or reduced at a narrowing point 30. A platinum catalyst 32 is packed in the large cross-sectional area of upper expansion zone 26 above the narrowing point 30 to maximize the contact area between the sample and the catalyst surfaces, and thus, minimize localized cooling, while in the small cross-sectional area of lower end 28, the platinum catalyst is packed in order to enhance oxidation. In the embodiment illustrated, the catalytic oxidation tube 12 has a wall thickness of 1.5 millimeters ("mm") and is approximately 260 mm in length with the upper expansion zone approximately 110 mm in length and the narrowed cross-sectional area approximately 150 mm in length. The outside diameter at the upper end of the catalytic oxidation tube is approximately 22 mm, while the outside diameter near an outlet end 34 of the narrowed cross-sectional area 28 is approximately 11 mm. In operation, oxygen is supplied from the oxygen source 20 to the catalytic oxidation tube 12 at a flow rate of approximately 200 milliliters per minute with

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the tube heated to approximately 850°C. All nitrogen compounds are quickly oxidized in the catalytic oxidation tube to NO/NO₂ according to the following equation:



5 The ratio of NO and NO₂ is determined by the type of nitrogen species, oxygens partial pressure, and effective temperature. In the embodiment illustrated, a 40 micro liter sample is used.

Referring back to Figure 1, the product gas, water vapor and carrier gas are discharged from the catalytic oxidation tube 12 through the outlet
10 34 and carried by a suitable line 36 to a condenser 38. The condenser 38 removes a substantial amount of water vapor from the product gas discharged from the catalytic oxidation 12.

From the condenser 38, NO and NO₂ and the carrier gas, oxygen, flows through a line 39 and into the N-N reaction chamber 14 where the NO₂ in
15 gaseous phase is converted in the presence of a reagent, described below, according to the following equation:

V(III)



It should be noted that since NO₂ is soluble in water, nitric acid
20 (HNO₃) formed in the condenser 38 is also provided to the reaction chamber 14 for conversion to NO.

The N-N reaction chamber 14 is shown in detail in Figure 3. The N-N reaction chamber 14 comprises a glass vessel 40 partially filled with an aqueous reagent 41 containing vanadium (III) and sulfuric acid (H₂SO₄) (Figures
25 4A and 4B). The reagent may comprise 0.1% to 5% vanadium-chloride by volume in water, while the sulfuric acid may comprise 5% to 60% by volume in water. Preferably, the reagent 41 comprises two percent vanadium-chloride and twenty percent sulfuric acid by volume in water. The N-N reaction chamber

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14 includes a first inlet 42 for the reagent and the product output from the catalytic oxidation tube 12. The aqueous reagent 41 is maintained between 60°C and 90°C, preferably at 80°C, while a flow of oxygen from the catalytic combustion tube 12 continuously sparges the reaction chamber 14.

5 The N-N reaction chamber 14 includes a second inlet 44 for injection of a second sample into the N-N reaction chamber with a microsyringe 43. The second sample is used to determine Combined Nitrate-Nitrite Nitrogen Content. The N-N reaction chamber 14 has an inner sleeve 46, spaced apart and generally shaped to conform to a lower portion 48 of the N-N reaction
10 chamber 14. The inner sleeve 46 is partially submerged in the reagent 41 (Figure 4A) with the submerged portion having holes 50 to allow gas and reagent 41 to pass therethrough. As illustrated in Figure 3, the inner sleeve 46 is sealed annularly to an inner wall 52 of the N-N reaction chamber below the reagent and carrier gas inlet 42, whereas the inner sleeve is sealed to the N-N
15 reaction chamber wall above an outlet 54. Since the inner sleeve 46 is sealed annularly to the inner wall 52, the oxygen carrier gas and product output from the condenser 38 flows through the N-N reaction chamber 14 by passing through the holes 50 in the inner sleeve 46. Preferably, the N-N reaction chamber 14 is made of Pyrex and is concentric having an outside diameter approximately 25
20 mm, while the inner sleeve 46 is positioned within the N-N reaction chamber 14 and has an outside diameter of approximately 15 mm. The N-N reaction chamber 14 has sufficient cross-sectional area such that when the sample is injected into the reagent from the microsyringe 43, the second sample falls into the reagent 41 without contacting the sides of inner sleeve 46. In the
25 embodiment as shown, quick reaction with the reagent 41 is provided by minimizing the reagent volume, which is approximately 6 milliliters. In the preferred embodiment, the N-N reaction chamber 14 is mounted in a heated aluminum block 51.

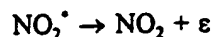
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When operated to determine Combined Nitrite-Nitrate Nitrogen Content, the aqueous nitrogen containing sample is injected into inlet 44 with the microsyringe 43 to fall to the reagent solution 41, as illustrated in Figure 4A. When the inlet 44 is closed, as illustrated in Figure 4B, pressure from the oxygen carrier gas through the inlet 42 pressurizes the N-N reaction chamber 14 such that the reagent is forced upward between the inner sleeve 46 and the inner wall 48. Upward movement of the reagent solution 41 is caused by flow of the carrier and product gas through the inlet holes 50. The N-N reaction chamber 14 reduces nitrate nitrogen and/or nitrite nitrogen in the sample to NO. The NO is sparged out from the reagent into the oxygen carrier gas through the outlet 54. A container 35 containing additional reagent and a pump 37 are connected to the N-N reaction chamber 14, for example, through line 39. The container 35 and the pump 37 supply additional reagent to the N-N reaction chamber 14 as needed.

Referring back to Figure 1, the outlet port 54 from the N-N reaction chamber 14 is connected to the drying means 16 comprising a condenser 60, a gas/liquid separator 62 and a dryer 64. The NO and the oxygen carrier gas flows from the outlet 54 of the N-N reaction chamber 14 to the condenser 60. The condenser 60 condenses liquid present in the NO and oxygen carrier gas from the N-N reaction chamber 14. A suitable fan 63 can be provided to aid in condensation of water vapor from the NO and oxygen carrier gas. The condenser 60 is further connected to the gas/liquid separator 62. The gas/liquid separator 62 separates the liquid condensed by condenser 60. A drain outlet 65 of the gas/liquid separator 62 is provided to discharge continuously the separated liquid. An outlet 66 from the gas/liquid separator 62 is connected to the dryer 64. Preferably, the dryer 64 is a permeation dryer having an inlet 70 connected to an oxygen supply 72 and a ventilation port 74. Since the dryer 64 principally functions to remove water vapor from, the oxygen carrier gas and NO flowing

from the liquid/gas separator 62, a second condenser 76 similar to condenser 60, can be coupled between the liquid/gas separator 62 and the dryer 64 to insure liquid water does not enter the dryer 64. An outlet 78 of the dryer 64 is connected through suitable tubing 80 as one input to a reaction chamber 82 of the chemiluminescent detector apparatus 18. The drying means 16 functions to receive a nitric oxide and oxygen mixture, dries the mixture by removal of water vapor, and lowers the dew point of the carried gases below a predetermined temperature level, a level that has been determined should not exceed the operating temperature of the reaction chamber 82. Oxygen from the oxygen source 72 is also supplied through tubing 86 to an ozone generator 88. The ozone generator 88 generates a supply of ozone which is applied through tubing 90 as a second input to the reaction chamber 82.

In the reaction chamber 82, the mixing of NO with ozone causes a reaction that forms metastable or excited nitrogen dioxide (NO_2^*). Almost instantaneously, the excited nitrogen dioxide relaxes to its ground state with a resulting photo-emission (ϵ), the following reactions occurring:



The detector module 18 detects the light emission (ϵ) which is proportional to the amount of NO, which in turn is proportional to the nitrogen content of the sample. Depending on whether the aqueous sample was injected into the system at the oxidation tube 12 or at the N-N reaction chamber, the amount of NO detected will be proportional to the Combined Total Nitrogen Content or the Combined Nitrate-Nitrite Nitrogen Content, respectively. An ozone scrubber 94 is connected to the reaction chamber and removes any remaining ozone upon completion of the reaction before venting the nitrogen dioxide through an outlet 96.

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In the embodiment illustrated in Figure 1, the various modules and units identified in the schematic block diagram can conveniently be comprised of the following commercially available equipment:

	Catalytic Oxidation Tube 12	P/N 888-000*
5	Condenser 38	P/N 888-205*
	N-N Reaction Chamber 14	P/N 888-379*
	Condenser 60	P/N 888-205*
	Dryer 64	Model MD-250-24 Permapure Products, Inc. Farmingdale, NJ.
10		
	NO/NO _x Detector 18	P/N 519-420*
15	* - Available from Rosemount Analytical Inc., Dohrmann Division, Santa Clara, CA.	

The gas/liquid separator 62 is of conventional design. For example, the gas/liquid separator 62 can be similar in shape to the gas/liquid separator (P/N 512-100) available from Rosemount Analytical Inc., Dohrmann
20 Division, Santa Clara, CA. However, the distance 67 between the drain 65 and the lower level of the liquid must be sufficient to accommodate the operating pressure of the present system.

The nitrogen detection system 10 illustrated in Figure 1 operates with positive pressure from the oxygen source 20 to carry the product gases
25 throughout the system. Alternatively, a vacuum pump 98 can be connected to the system to increase sensitivity by driving the product gases through the system.

In summary, the present invention provides a convenient and versatile system for measuring Total Combined Nitrogen Content and Combined
30 Nitrate-Nitrite Nitrogen Content in an aqueous sample. The Total Combined Nitrogen Content is measured by pyrolyzing the sample in a catalytic oxidation

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tube, the output of which is connected to an N-N reaction chamber. The N-N reaction chamber converts NO_2 to NO with a liquid chemical reduction agent comprising vanadium and sulfuric acid that is then mixed in a reaction chamber with ozone to form metastable nitrogen dioxide, which then relaxes to its ground state with a resulting photo-emission is measured by a detector such as a chemiluminescent detector. The chemiluminescent detector provides a signal proportional to the Total Combined Nitrogen content contained in the aqueous sample.

In addition, the present invention can be used to determine the combined Nitrate-Nitrite Nitrogen Content in the aqueous sample by injecting a second sample directly into the N-N reaction chamber containing the same liquid chemical reduction agent, the resulting NO produced being carried to and measured by the detector, as previously described. From these determinations, the Total Kjeldahl Nitrogen can be calculated as the difference between the Total Combined Nitrogen Content and the Combined Nitrate-Nitrite Nitrogen Content.

Although the present invention has been described with reference to preferred embodiments, workers skilled in the art will recognize that changes may be made in form and detail without departing from the spirit and scope of the invention.

WHAT IS CLAIMED IS:

1. An apparatus for determining the Total Nitrogen Content and Combined Nitrate-Nitrite Nitrogen Content in a liquid system, the apparatus comprising:

an oxygen source providing a carrier gas;
oxidation means connected to the oxygen source and receiving the carrier gas and a first sample extracted from the liquid system, the oxidation means having an oxidation catalyst and converting the first sample to a product gas including NO_2 ;

reaction means connected to the oxidation means to receive the carrier gas, the reaction means having a liquid chemical reduction agent and converting the NO_2 to NO ;

detection means connected to the reaction means to receive the NO , the detection means measuring the amount of NO to determine the Total Nitrogen Content in the liquid system;
and

wherein the reaction means receives a second sample extracted from the liquid system, the second sample having at least one of nitrites and nitrates and the reaction means converting at least one of the nitrites and nitrates to a second quantity of NO , and wherein the detection means receives and measures the second quantity of NO to determine Combined Nitrate-Nitrite Nitrogen Content in the liquid system.

2. The apparatus as specified in claim 1 wherein the liquid chemical reduction agent comprises vanadium (III) and sulfuric acid.

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3. The apparatus as specified in claim 1 wherein the liquid chemical reduction agent comprises approximately 0.1% to 5% vanadium chloride by volume and approximately 5% to 60% sulfuric acid by volume.
4. The apparatus as specified in claim 3 wherein the liquid chemical reduction agent comprises approximately 2% vanadium chloride by volume and approximately 20% sulfuric acid by volume.
5. The apparatus as specified in claim 1 wherein the liquid chemical reduction agent is maintained at a temperature between 60°C and 90°C.
6. The apparatus as specified in claim 1 wherein the detection apparatus comprises a chemiluminescent detector.
7. The apparatus as specified in claim 1 and further comprising vacuum means to pull the carrier gas and product gas to the detection apparatus.
8. The apparatus as specified in claim 1 wherein the carrier gas consists essentially of oxygen.
9. A method for determining Total Nitrogen Content and Combined Nitrate-Nitrite Nitrogen Content in a liquid system, the method comprising the steps:
 - extracting a first liquid sample and a second liquid sample from the liquid system;
 - introducing the first liquid sample with a carrier gas comprising oxygen into an oxidation means having an oxidation

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catalyst for converting the first liquid sample to a product gas comprising NO_2 ;
converting the product gas comprising NO_2 to a quantity of NO in a reaction means having a liquid chemical reduction agent;
measuring the quantity of NO with a detection apparatus to determine the Total Nitrogen Content in the liquid system;
introducing the second liquid sample with the carrier gas into the reaction means having the liquid chemical reduction agent for converting at least one of nitrates and nitrites in the second the liquid sample to a second quantity of NO ; and
measuring the second quantity of NO with the detection apparatus to determine the Combined Nitrate-Nitrite Content in the liquid system.

10. The method as specified in claim 9 wherein the carrier gas consists essentially of oxygen.

11. The method as specified in claim 9 wherein the liquid chemical reduction agent comprises vanadium (III) and sulfuric acid.

12. A method for determining Combined Nitrate-Nitrite Nitrogen Content in a liquid system, the method comprising the steps:
extracting a liquid sample from the liquid system;
introducing the liquid sample with a carrier gas including oxygen into a reaction means having a liquid chemical reduction agent comprising vanadium (III) and sulfuric acid and

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converting at least one of nitrates and nitrites in the liquid sample to NO; and
measuring the NO with a detection apparatus to determine the Combined Nitrate-Nitrite Content in the liquid system.

13. The method as specified in claim 12 wherein the Total Combined Nitrogen Content in the liquid system is determined, and wherein the step of extracting includes extracting a second liquid sample from the liquid system, and wherein the method further comprises:

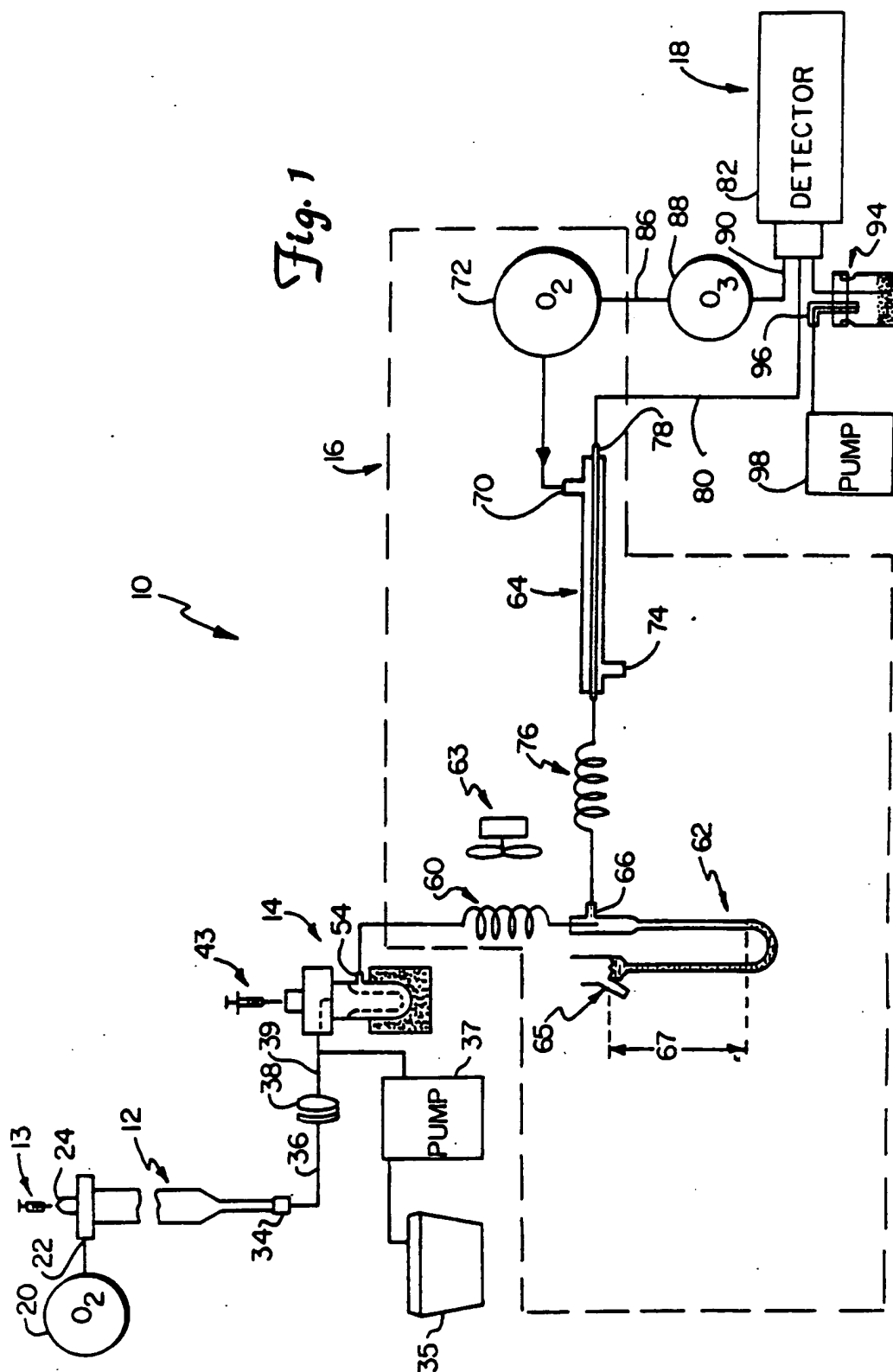
introducing the second liquid sample with the carrier gas including oxygen into an oxidation means having an oxidation catalyst and converting the second liquid sample to a product gas comprising NO₂

converting the product gas comprising NO₂ to a second quantity of NO in the reaction means having the liquid chemical reduction agent; and

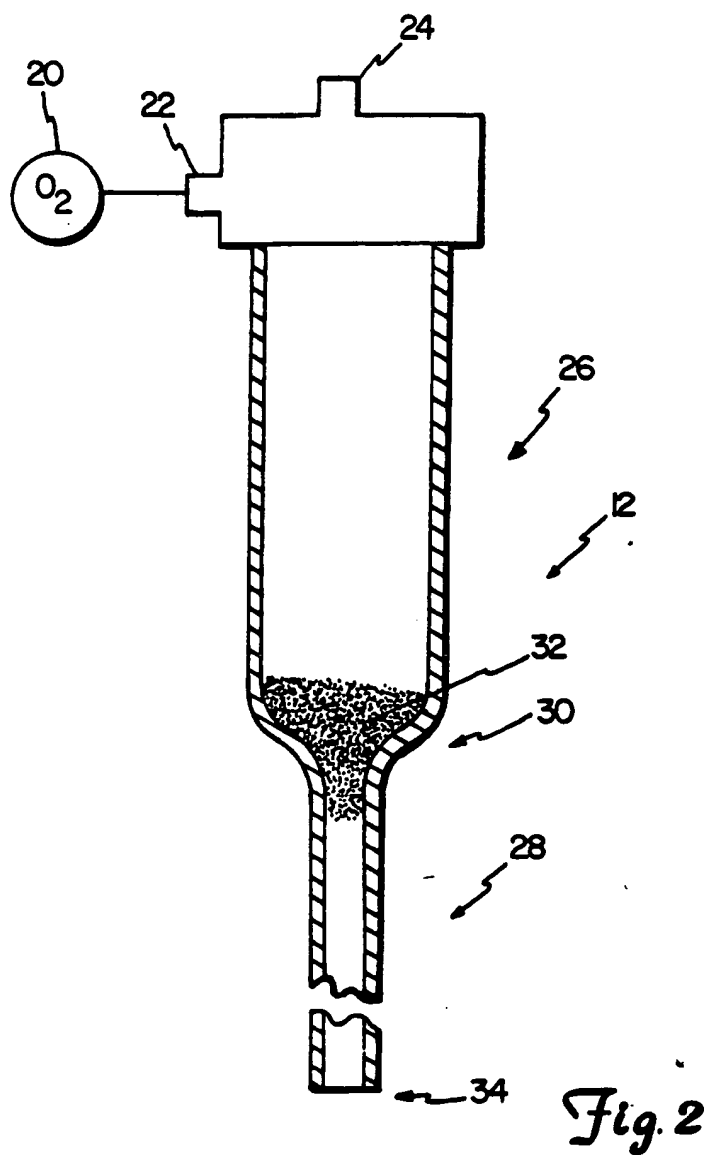
measuring the second quantity of NO with the detection apparatus to determine the Total Nitrogen Content in the liquid system.

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Fig. 1



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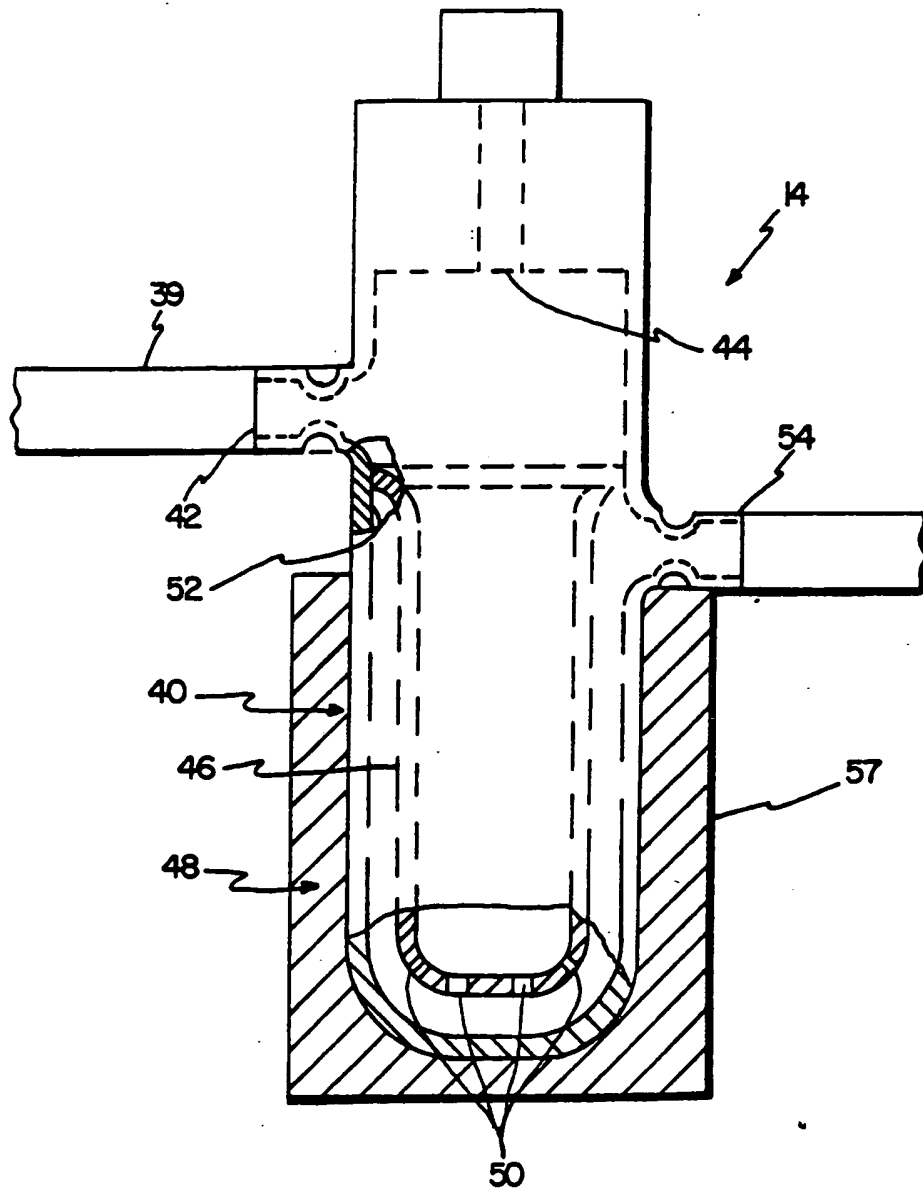


Fig. 3

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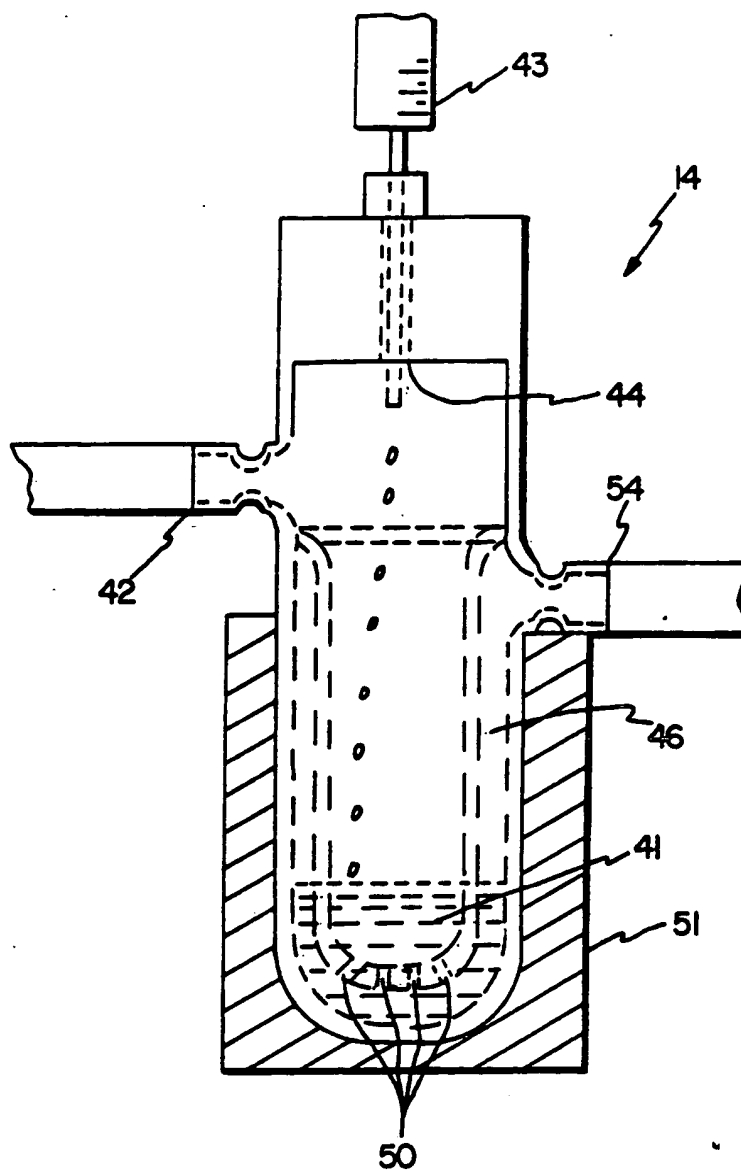


Fig. 4A

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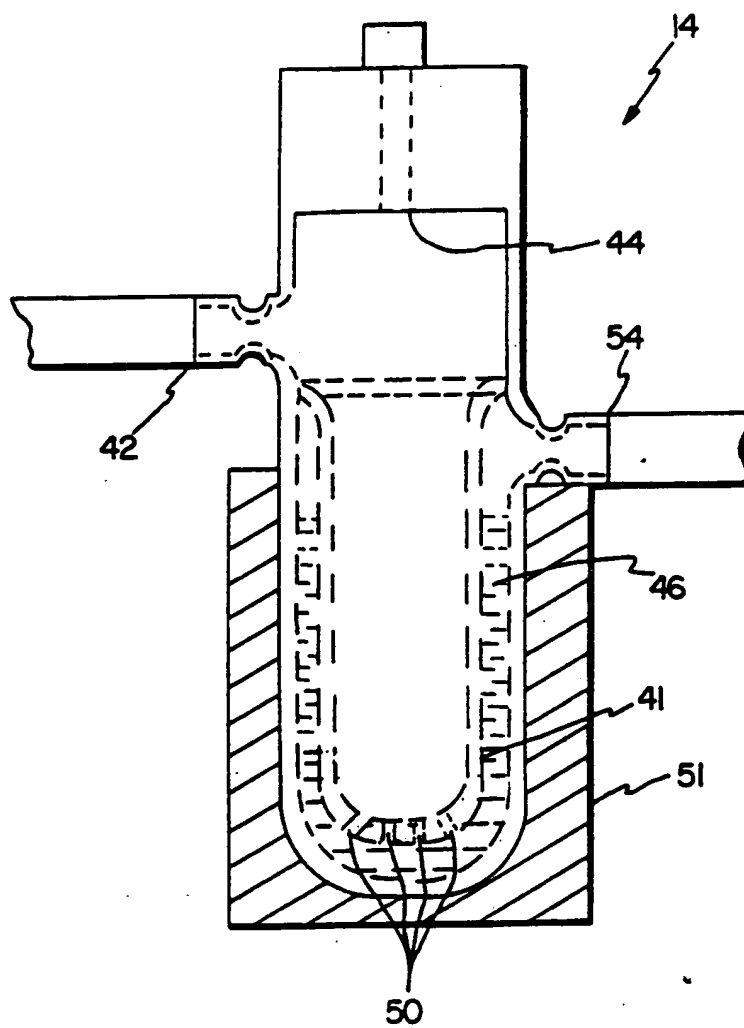


Fig. 4B

INTERNATIONAL SEARCH REPORT

International Application No.
PCT/US 93/08593

A. CLASSIFICATION OF SUBJECT MATTER
IPC 5 G01N31/00 G01N31/12

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)
IPC 5 G01N

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practical, search terms used)

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
Y	US,A,3 877 875 (R.H.JONES; M.B.JOHNSTON) 15 April 1975	1,6-10
A	see the whole document see particularly column 5, lines 28-34 and lines 50-57;column 6,lines 6-42. ---	2-5, 11-13
Y	PATENT ABSTRACTS OF JAPAN vol. 11, no. 2 (P-532)(2449) 6 January 1987 & JP,A,61 180 139 (MITSUBISHI HEAVY IND LTD) 12 August 1986	1,6-10
A	see abstract ---	2-5,11, 12
A	US,A,4 018 562 (R.E.PARKS; R.L.MARIETTA) 19 April 1977 cited in the application ---	1,9,12
-/--		

☒ Further documents are listed in the continuation of box C.

☒ Patent family members are listed in annex.

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Date of the actual completion of the international search

6 January 1994

Date of mailing of the international search report

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Name and mailing address of the ISA

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INTERNATIONAL SEARCH REPORT

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C.(Continuation) DOCUMENTS CONSIDERED TO BE RELEVANT		
Category	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
A	FR,A,2 415 299 (SUMITOMO CHEMICAL COMPANY LTD) 17 August 1979 see the whole document ---	1,9,12
A	EP,A,0 020 072 (SUMITOMO CHEMICAL COMPANY) 10 December 1980 see the whole document ---	1,9,12
A	GB,A,2 001 045 (NV PHILIPS' GLOEILAMPENFABRIEKEN) 24 January 1979 see the whole document ---	1,9,12
A	EP,A,0 075 467 (SUMITOMO CHEMICAL COMPANY LTD) 30 March 1983 see the whole document -----	1,9,12

INTERNATIONAL SEARCH REPORT

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International Application No

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